Nonadiabatic dynamics and coherent control of nonequilibrium superconductors

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Workshop on strongly correlated electron systems

Schloss Ringberg, November 2013

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1. Introduction
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   - How does a quantum system relax?

2. Nonadiabatic dynamics of superconductors
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   - Density matrix formalism

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   - Generation of coherent phonons in non-equilibrium SC state:
     beating phenomena, resonant generation of coherent phonons
   - Experimental signatures: Oscillations of pump-probe conductivity

4. Conclusions & Outlook
Motivation I: Recent pump-probe experiments

- THz-pump pulse: Excite with intense femto-second pulse, induce dynamics
- THz-probe pulse: After delay time $\delta t$, measure with second, less intense pulse

Optical conductivity / transmittivity as a function of $\omega$ and $\delta t$ gives information about:

- Dynamics of SC condensate, order parameter oscillations
- Cooper pair recombination / recovery dynamics
- Coherent phonon oscillations

SC gap $\Delta$

$\sim 1 - 10$ meV

$\Delta = 4.1$ meV

$\Delta = 1$ ps$^{-1}$

$\Delta = 33$ cm$^{-1}$
Motivation I: Recent pump-probe experiments

**THz-pump THz-probe spectroscopy on NbN films**

- pump-pulse duration: 90 fs

**non-adiabatic** excitation of SC:

$$\tau_p \ll \tau_\Delta \sim \hbar/(2|\Delta|)$$

- Measure change in transmission of probe field $$\delta E$$

**Observation:**

- algebraically damped oscillations in $$\delta E$$ as a function of delay time $$\delta t$$

- frequency changes with laser intensity

**Interpretation:**

order parameter amplitude oscillations
Motivation II: How does a quantum system relax?

How does a quantum system thermalize?

As \( t \to \infty \), generic observables \( \hat{O} \) become time independent.

- **True relaxation vs. decoherence:**
  - **Decoherence:** Only averaged observables become time-independent.
  - **True relaxation:** Generic *local* (unaveraged) observables become time-independent.

  \[
  \tilde{O} = \lim_{t \to \infty} \langle \Psi(t) | \hat{O} | \Psi(t) \rangle \quad \text{time independent}
  \]

  - (i) systems coupled to bath, (ii) closed systems with certain interactions (infinite closed systems; otherwise recurrence)

**Open questions:**

- Which type of interactions lead to thermalization?
- Which observables \( \hat{O} \) to consider?
- How to describe thermalized state?
  (is there a density matrix (ensemble) \( \rho \) such that: \( \tilde{O} = \text{Tr}[\hat{O} \rho] \)?)
SC coupled to laser field and optical phonons

Goal: simulate non-adiabatic dynamics of superconductor coupled to
(i) pump laser field and (ii) optical phonons

Microscopic model:

\[
H = \sum_{k} \epsilon_k c_{k,s}^+ c_{k,s} + \sum_{k} \left( \Delta_k c_{k,\uparrow}^+ c_{-k,\downarrow} + \text{c.c.} \right) \\
- \frac{e\hbar}{m} \sum_{kq} (k \cdot A_q) c_{k+q/2,s}^+ c_{k-q/2,s} + \frac{e^2}{2m} \sum_{kq} (A_{q-k} \cdot A_q) c_{k,s}^+ c_{k,s} \\
+ \sum_{q,j} \hbar \omega_{q,j} \left( b_{q,j}^+ b_{q,j} + \frac{1}{2} \right) + \sum_{p,j,k,s} \left( g_{pjk}s (b_{-p,j}^+ + b_{p,j}^+) c_{k+p,s}^+ c_{k,s} + \text{c.c.} \right)
\]

with gap equation: \( \Delta_k = \sum_{k'} V_{kk'} \langle c_{-k',\downarrow} c_{k',\uparrow} \rangle \)

Gaussian pump and probe pulse: \( A_q(t) = A_0 e^{-(t/\tau)^2} \left( \delta_{q,q_0} e^{-i\omega_0 t} + \delta_{q,-q_0} e^{+i\omega_0 t} \right) \)

pump pulse: \( \hbar \omega_0 \gtrsim 2|\Delta| \)

probe pulse: \( \hbar \omega_0 \lesssim 2|\Delta| \)
Theoretical methods to compute non-equilibrium dynamics in SCs

**Goal:** simulate *non-adiabatic* dynamics of superconductor coupled to optical phonons

→ considered *hierarchies of time-scales:* \( \tau_p \ll \tau_{ph}, \tau_\Delta \ll \tau_\epsilon \)

- *time-dependent Ginzburg Landau theory, mu*-*, and T*- models:*
  - quasi-equilibrium is assumed at all times
  - time evolution of a single collective order parameter
  - only valid for: \( \tau_\Delta \gg \tau_\epsilon \) (i.e., close to \( T_c \), very dirty SCs)

- *Boltzmann kinetic equation:*
  - requires adiabaticity on all time-scales
  - does not capture coherent evolution of quasi-particle distributions
  - only valid for: \( \tau_p \gg \tau_\Delta, \tau_{ph} \)

- *Phenomenological Rothwarf-Taylor models:*
  - rate equations for quasi-particle and phonon occupations
  - only valid for: \( \tau_p \gg \tau_\Delta, \tau_{ph} \)

→ instead use density matrix formalism (expansion in \( \sim g_{ph} t / \hbar \))

\[
\frac{d}{dt} \mathcal{O} = \frac{i}{\hbar} [H, \mathcal{O}]
\]
Density matrix formalism: Equations of motion

Superconducting state: Bogoliubov transformation

\[ \alpha_k^\dagger = u_k c_k^\dagger + v_k^* c_{-k} \]
\[ \beta_k^\dagger = u_k c_{-k} - v_k c_k \]
\[ \Delta_k = \sum_{k'} W_{k,k'} \left[ u_{k'} v_{k'} \left( 1 - \langle \alpha_{k'}^\dagger \alpha_{k'} \rangle - \langle \beta_{k'}^\dagger \beta_{k'} \rangle \right) + u_{k'}^2 \langle \beta_{k'} \alpha_{k'} \rangle - v_{k'}^2 \langle \alpha_{k'}^\dagger \beta_{k'}^\dagger \rangle \right] \]

All quantities of interest can be expressed in terms of these dynamical variables

\[ \langle \alpha_{k,k'} \rangle (t), \quad \langle \beta_{k,k'} \rangle (t), \quad \langle \alpha_{k,k'}^\dagger \rangle (t), \quad \langle \alpha_k \beta_{k'} \rangle (t), \quad \langle b_p \rangle (t), \quad \langle b_p^\dagger \rangle (t) \]

**Current density:**

\[ j(q,\omega) \simeq \frac{e\hbar}{mV} \sum_k k \left[ \langle \alpha_{k,k+q}^\dagger \alpha_{k+q} \rangle - \langle \beta_{k+q,k}^\dagger \beta_k \rangle + k \cdot q \frac{\hbar^2 |\Delta_1|}{2m (E_k^2)} \left( \langle \alpha_{k,k+q}^\dagger \beta_{k+q} \rangle + \langle \alpha_{k+q,k} \beta_k \rangle \right) \right] \]

**Lattice displacement:**

\[ U(r,t) = \sqrt{\frac{\hbar}{2M\omega_{ph}V}} \sum_p D_p(t) e^{i p \cdot r} \]

with the **coherent phonon amplitude:**

\[ D_p(t) = \langle b_p \rangle + \langle b_p^\dagger \rangle \]

Density-matrix theory:

\[ \frac{d}{dt} \mathcal{O} = \frac{i}{\hbar} [H, \mathcal{O}] \]

\[ \Rightarrow \text{ yields equations of motions for the above expectation values} \]
Density matrix formalism: Equations of motion

Density-matrix theory: \[ \frac{d}{dt} \mathcal{O} = \frac{i}{\hbar} [H, \mathcal{O}] \]

\( \Rightarrow \) yields equations of motions for

\[ \langle \alpha_{k}^\dagger \alpha_{k'} \rangle (t), \quad \langle \beta_{k}^\dagger \beta_{k'} \rangle (t), \quad \langle \alpha_{k}^\dagger \beta_{k'}^\dagger \rangle (t), \quad \langle \alpha_{k} \beta_{k'} \rangle (t), \quad \langle b_{p} \rangle (t), \quad \langle b_{p}^\dagger \rangle (t) \]

For example

\[ i\hbar \frac{d}{dt} \langle \alpha_{k}^\dagger \beta_{k+q}^\dagger \rangle = - (R_{k} + R_{k+q}) \langle \alpha_{k}^\dagger \beta_{k+q}^\dagger \rangle + C_{k+q}^* \langle \alpha_{k}^\dagger \alpha_{k+q} \rangle + C_{k}^* \left( \langle \beta_{k+q}^\dagger \beta_{k} \rangle - \delta_{q,0} \right) \]

\[ + \frac{e\hbar}{2m} \sum_{q' = \pm q_{0}} 2\mathbf{k} \cdot A_{q'}(t) \left\{ - L_{k+q}^+ \langle \alpha_{k+q}^\dagger \beta_{k+q}^\dagger \rangle + L_{k+q,-q'}^+ \langle \alpha_{k}^\dagger \beta_{k+q-q'}^\dagger \rangle \right. \]

\[ - M_{k+q,-q'}^- \langle \alpha_{k}^\dagger \alpha_{k+q-q'} \rangle + M_{k,q'}^- \left( \langle \beta_{k+q}^\dagger \beta_{k+q'} \rangle - \delta_{q,q'} \right) \left\} + \cdots \]

Up to order \( (A_{q})^n \), laser field only couples \( (k, k') \) to \( (k, k' + nq) \)

\( \Rightarrow \) leads to an effectively one-dimensional system of equations

\( \Rightarrow \) interaction with laser field can be computed essentially exactly.

[\textit{Papenkort, Kuhn, Axt, PRB 08}]
Response of superconductor (w/o phonons)

- Two regimes:
  - Adiabatic behavior for $\tau_\Delta \ll \tau_p$
  - Non-adiabatic behavior for $\tau_p \ll \tau_\Delta$

- Algebraically damped order parameter oscillations after short pump pulse ($\tau_p \ll \tau_\Delta$):

$$|\Delta| = |\Delta_\infty| + \Gamma \frac{\cos\left(\frac{2|\Delta_\infty|}{\hbar} t + \Phi\right)}{\sqrt{t}}$$

("Higgs" amplitude mode)

[Volkov, Kogan, JETP 74]
[Yuzbashyan, Altshuler PRL 06]
Algebraically damped oscillations due to decoherence

- Algebraically damped oscillations in averaged quantities due to decoherence

\[
\Delta = W_0 \sum_{k \in \mathcal{W}} \langle c_{-k, \downarrow} c_{+k, \uparrow} \rangle
\]

\[
\dot{q}_0 = \frac{-e\hbar}{2mV} \sum_k (2k + q_0) \langle c_{k, \sigma}^\dagger c_{k+q_0, \sigma} \rangle
\]

- But no true relaxation!

Order parameter oscillations

Quasiparticle occupations

\[
\langle \alpha_k^\dagger \alpha_k \rangle
\]
Gap oscillations: Comparison with experiment

- Qualitative agreement between theory and experiment

Numerical simulations:

\[ |\Delta| = |\Delta_\infty| + \Gamma \cos(\frac{2|\Delta_\infty|}{\hbar} t + \Phi) \sqrt{t} \]

THz-pump THz-probe spectroscopy NbN:

\[ \tau_p = 60 \text{ fs} \]

Matsunaga, Shimano, et al.
PRL 111, 057002 (2013)
Response of superconductor (w/o phonons)

Pump-probe conductivity shows signatures of non-adiabatic dynamics

\[ \sigma(\delta t, \omega) = \frac{j(\omega)}{i\omega A(\omega)} \]

⇒ oscillations in pump-probe response as a function of delay time \( \delta t \)

Numerical simulation:

THz-pump THz-probe spectroscopy NbN:


Matsunaga, Shimano, et al.
PRL 111, 057002 (2013)
Superconductor coupled to optical phonons

- Linear coupling to optical phonons:

\[ H_{\text{el-ph}} = g_{\text{ph}} \sum_{\mathbf{p}, \mathbf{k}, \sigma} \left[ (b_{-\mathbf{p}}^{\dagger} + b_{\mathbf{p}}) c_{\mathbf{k}+\mathbf{p}, \sigma}^{\dagger} c_{\mathbf{k}, \sigma} + \text{c.c.} \right] \]

- Perform expansion in \( g_{\text{ph}} \) using \( \frac{d}{dt} \mathcal{O} = \frac{i}{\hbar} [H, \mathcal{O}] \)

\[ \Rightarrow \text{infinite hierarchy of equations of motion} \]

- Study generation of coherent phonons:

- Break hierarchy at first order: \( \langle \alpha_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k'}}^{\dagger} b_{\mathbf{p}} \rangle \approx \langle \alpha_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k'}}^{\dagger} \rangle \langle b_{\mathbf{p}} \rangle \)

- Non-vanishing \( \langle b_{\mathbf{p}} \rangle \) leads to finite lattice displacement:

\[ U(\mathbf{r}, t) = \sqrt{\frac{\hbar}{2M\omega_{\text{ph}}V}} \sum_{\mathbf{p}} D_{\mathbf{p}}(t)e^{i\mathbf{p}\cdot\mathbf{r}}, \]

with \( D_{\mathbf{p}} = (b_{-\mathbf{p}}^{\dagger} + b_{\mathbf{p}}) \)

- Equation of motion for coherent phonon amplitude:

with forcing term:

\[ \mathcal{F}_{\mathbf{p}}(t) = -\frac{2\omega_{\text{ph}}}{\hbar} g_{\text{ph}} \sum_{\mathbf{k}} \left[ M_{\mathbf{k}, \mathbf{p}}^{+} \left( \langle \alpha_{\mathbf{k}+\mathbf{p}}^\dagger \beta_{\mathbf{k}} \rangle - \langle \alpha_{\mathbf{k}}^\dagger \beta_{\mathbf{k}+\mathbf{p}}^\dagger \rangle \right) + L_{\mathbf{k}, \mathbf{p}}^{-} \left( \langle \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}+\mathbf{p}} \rangle + \langle \beta_{\mathbf{k}+\mathbf{p}}^\dagger \beta_{\mathbf{k}} \rangle \right) \right] , \]
Equation of motion for coherent phonon amplitude

- Equation of motion for coherent phonon amplitude:

\[
\left[ \frac{d^2}{dt^2} + \omega_{\text{ph}}^2 \right] D_p(t) = \mathcal{F}_p(t), \quad \mathcal{F}_p(t) = -\frac{2\omega_{\text{ph}}}{\hbar} g_{\text{ph}} \sum_k \left[ M_{k,p}^+ \left( \langle \alpha_{k+p} \beta_k \rangle - \langle \alpha_k \beta_{k+p}^\dagger \rangle \right) + \cdots \right]
\]

\[\text{Hierarchy of time scales: } \tau_p \ll \tau_\Delta \ll \tau_{\text{ph}}\]

\[\Rightarrow \text{Forcing term can be approximated by } \mathcal{F}_p(t) \simeq A_p \Theta(t)\]

\[\Rightarrow D_p(t) \simeq \frac{A_p}{\omega_{\text{ph}}^2} \left[ 1 - \cos(\omega_{\text{ph}} t) \right]\]

\[\text{Displacive excitation of coherent phonons: } \]

(similar to semiconductors)

- abrupt change in quasiparticle states leads to jump in equilibrium position of lattice
- cosine oscillations in lattice displacement
- extrema at integer and half-integer values of \( \tau_{\text{ph}} \)
Generation of coherent phonons: Numerical results

Hierarchy of time scales: \( \tau_p \ll \tau_\Delta \ll \tau_{ph} \) \( \Rightarrow \) \( D_p(t) \simeq \frac{A_p}{\omega_{ph}^2} \left[ 1 - \cos(\omega_{ph} t) \right] \)

- **Numerical results for displacive excitation of coherent phonons:**

![Graph showing cosine oscillations and extrema](image)

- **cosine oscillations** in lattice displacement \( U(R_0, t) \)
- **extrema at integer and half-integer values of** \( \tau_{ph} \)

**Parameter values**
- \( \tau_p = 0.05 \text{ ps}, \ h\omega_{ph} = 0.05 \text{ meV} \)
- \( \tau_p = 0.05 \text{ ps}, \ h\omega_{ph} = 0.10 \text{ meV} \)
- \( \times 50 \) \( \tau_p = 2.00 \text{ ps}, \ h\omega_{ph} = 0.05 \text{ meV} \)
- \( \times 50 \) \( \tau_p = 2.00 \text{ ps}, \ h\omega_{ph} = 0.10 \text{ meV} \)
- \( \times 400 \) \( \tau_p = 10.0 \text{ ps}, \ h\omega_{ph} = 0.05 \text{ meV} \)
- \( \times 400 \) \( \tau_p = 10.0 \text{ ps}, \ h\omega_{ph} = 0.10 \text{ meV} \)

**References**
- APS, Manske, Avella, PRB 84, 214513 (2011)
Generation of coherent phonons: Quantum beats

Hierarchy of time scales: \[ \tau_p \ll \tau_\Delta \sim \tau_{ph} \]

⇒ forcing term can be approximated by \[ \mathcal{F}_p(t) \simeq \Theta(t)[A_p + B_p \cos(2\Delta_\infty t/\hbar) / \sqrt{t}] \]

⇒ quantum beating when \[ \omega_d = |\omega_\Delta_\infty - \omega_{ph}| \ll \omega_{ph} \]

⇒ \[ D_p(t) \simeq \frac{B_p}{\omega_{ph}} \sqrt{\frac{\pi}{2\omega_d}} [\cos(t\omega_{ph})S_2(t\omega_d) + \sin(t\omega_{ph})C_2(t\omega_d)] \]
Generation of coherent phonons: Pump-probe conductivity

Hierarchy of time scales: $\tau_p \ll \tau_\Delta \sim \tau_{ph}$

Signature of non-adiabatic dynamics in pump-probe conductivity

#### (a) Re($\sigma$) [arb. units]

- beating phenomenon in pump-probe conductivity $\sigma(\delta t, \omega = \omega_{ph})$

#### (b) Re[$\sigma(\omega = \omega_{\Delta})$]

#### (c) FT-Amp.

#### (d) |$\Delta$| [meV]

#### (e) $U(0,t)$ [arb. units]

Generation of coherent phonons: Resonance

Hierarchy of time scales: \( \tau_p \ll \tau_{\Delta} = \tau_{\text{ph}} \)

\[ \Rightarrow \text{resonant generation of coherent phonons for } \omega_{\text{ph}} = \omega_{\Delta \infty} (= \frac{2 \Delta_{\infty}}{\hbar}) \]

\[ \Rightarrow D_p(t) \approx \frac{B_p}{\omega_{\text{ph}}} \sqrt{t} \sin(\omega_{\text{ph}} t) + \cdots \]

Lattice displacement:

pump-probe conductivity:

resonantly enhanced oscillations in pump-probe conductivity as a function of delay time \( \delta t \)
Conclusions & Outlook

▶ Microscopic simulation of ultrafast dynamics in superconductors

▶ Non-adiabatic regime $\tau_p \ll \tau_\Delta, \tau_{ph}$:
  - order parameter oscillations
  $\Rightarrow$ qualitative agreement with experiment
  - generation of coherent phonons
  - for $\hbar \omega_{ph} = 2\Delta_\infty$: resonant enhancement of coherent phonons

▶ Pump-probe conductivity:
  - oscillations in $\sigma(\delta t, \omega)$ as a function of delay time
    with frequencies $\omega_{\Delta\infty} = 2\Delta_\infty/\hbar$ and $\omega_{ph}$
  - strong enhancement of oscillation amplitude
    when frequencies are in resonance: $\omega_{\Delta\infty} \approx \omega_{ph}$

▶ Outlook:
  - consider higher order in correlation expansion:
    incoherent phonons, feedback on SC condensate